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Nanofiltration of bulk drug industrial effluent using indigenously developed functionalized polyamide membrane

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HIGHLIGHTS

- Development of 4-layered indigenous functionalized nanofiltration membrane.
- Processing of bulk drug industrial effluent by nanofiltration.
- Development of mathematical model and simulation program using statistical mechanical equations.
- Scale-up and economic estimation for comparison of NF and RO systems.

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ABSTRACT

Nanofiltration (NF) and reverse osmosis (RO) and are membrane-based separation processes which can be used for treatment of industrial effluents and wastewater recycling. In the present study, performance of functionalized nanofiltration (FNF-400) and thin film composite (TFC) polyamide RO membranes has been investigated for the treatment of industrial effluent consisting of 5710 ppm total dissolved solids (TDS), 4050 ppm chemical oxygen demand (COD) and 8.4 ppm biochemical oxygen demand (BOD). Effect of various parameters such as applied pressure and feed composition on parameters such as permeate conductivity and flux, TDS rejection, turbidity removal besides COD and BOD reduction was evaluated. At a constant feed pressure of 21 bar, higher average flux of 36.95 L/m^2 h was observed in case of NF when compared to 18.77 L/m^2 h for RO. The % rejections of TDS, turbidity and COD were observed to be 85%, 97.8% and 73.33% for NF and 95%, 100% and 86.66%, respectively for RO systems. A statistical mechanical model was developed for commercial NF and RO systems. The economic estimation of commercial NF/RO systems was carried out which reveals that NF process is more economical due to lower operating pressures and consequently lesser energy consumption.

1. Introduction

High pressure membrane processes such as NF and RO are increasingly used for desalination of seawater, especially in coastal regions to combat high salinity in brackish water and water scarcity. In many regions ground water quality is one of the important factors for which NF/RO membranes have been applied for the production of safe and clean drinking water at low operating pressure with high water flux [1]. There is a growing awareness on the importance of trace levels of contaminants such as endocrine disrupting compounds (EDCs), pharmaceutically active compounds (PhACs), fluorides and dyes originating from industrial, agricultural, medical and domestic processes [2,3]. Compounds used in pharmaceuticals, personal care products and other consumables may enter aquatic environments after passing though wastewater treatment plants (WWTP), which are often not designed to remove such chemicals [4–6]. An integrated understanding of organic micro pollutant rejection mechanisms by NF/RO membranes has put the perspective on solutemembrane interactions which are influenced by process conditions and feed water composition [7,8].

A key component in a successful sustainable and economical implementation of NF/RO systems in wastewater polishing is to find a disposal or reuse strategy for the resulting concentrates/rejects [9,10]. Concentrates from NF process cannot be discharged directly into water bodies due to legislation and environmental regulations. For any discharge concept, apart from dilution to meet discharge levels, it is imperative to minimize the concentrate volume [11]. Minimization of the concentrate is often hampered by severe fouling effects with the main fouling constituents in the effluent being biologically inert dissolved organics originating from the WWTP influent and biological residue products from the

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Nomenclature

A_w	water permeability co-efficient (m^3 m/m^2 s mm Hg)
C_p	concentration of the permeate (mol/m^3)
C_f	concentration of the feed (mol/m^3)
C_W	membrane wall concentration (mol/m^3)
C_b	bulk solution concentration (mol/m^3)
$\Delta\Pi$	osmotic pressure (bar)
J_V	volumetric flux (L/m^2 h)
J_W	water flux (L/m^2 h)
U_i	transport velocity of species i and j (m/s)
$\nabla_{T\mu_i}$	equivalent isothermal gradient of chemical potential of species i
F_i	external force per mole on species i
ΔP	total pressure gradient (bar)
ΔP_m	trans membrane pressure (bar)
D_{ij}	diffusion coefficient between species i and j within the membrane (m^2/s)
D_{iM}	diffusion coefficient between species i and the membrane (m^2/s)
B_O	viscous flow parameter
H	viscosity of the solution in the membrane (kg/m s)
α_I	Dimensionless parameter
D_{ij}^T	multi-component thermal diffusion coefficient (m^2/s)
L_{ij}^C	Onsager coefficient in a center of mass frame of reference
L_o	viscous flow coefficient
α_1, α_2	coefficients that describe viscous separation effects
L_p	hydraulic conductivity (m/s)
Ω	coefficient of solute permeability
σ_v	reflection coefficients for volume flow
σ_s	reflection coefficient for solute flux
Pe	Peclet number
P	solute diffusive permeability (m/s)
Δz	effective thickness of the membrane (mm)
C_1	diffusion factor
C_2	selectivity factor
D_1	flow factor (mPa^{-1} s^{-1})
D_2	membrane constant (kg m^{-2} Pa^{-1} s^{-1})
R	universal gas constant (8.314 J K^{-1} mol^{-1})
T	absolute temperature (301 K).
n_i	the number of ions formed when the solute dissociates
C_i	the TDS concentration (mol L^{-1})

activated sludge process [12]. To overcome these problems, several researchers have recently focused their work on the removal of organic micro pollutants, EDCs, PhACs, fluoride and dyes using NF/RO membrane technology [13].

Amjad [14] tested membrane-based separation processes like ultrafiltration (UF) and reverse osmosis (RO) for treating a wide variety of industrial effluents. Jain et al. [15] used NF and RO processes for the treatment of leather plant effluent. From their studies, BOD and COD values of treated effluent were found to be well within the permissible limits. Tinghui et al. [16] reported rejection values of ⩾90% for most types of ionic compounds by RO membranes. Erswell et al. [17] used NF membranes to retain organic compounds of low molecular weight such as hydrolyzed reactive dyes as well as dyeing auxiliaries, divalent ions and certain large monovalent ions.

In the present investigation, an attempt was made to compare the efficiency of NF and RO processes for the treatment of bulk drug industrial effluent. Color, TDS, conductivity, turbidity, COD and BOD contents present in the feed, permeate and reject samples were analyzed by standard methods. The effect of various operating conditions on water flux and % rejection has been evaluated. With the obtained results, a mathematical model based on statistical mechanical transport equations was developed for commercial RO system. A detailed economic estimation of commercial NF/RO systems is presented.

2. Experimental work

2.1. Materials

Indigenously synthesized membranes of TFC polyamide RO and functionalized polyamide (FNF-400) were scaled-up to spiral wound membrane modules having effective separation area of 2.5 m^2 each, with the help of facilities available with Permionics Membranes Pvt. Ltd., Vadodara, India. The bulk drug industrial effluent was obtained from GVK Bio Pharma Ltd., IDA Nacharam, Hyderabad, India and the respective characteristics are depicted in Table 1. Potassium dichomate, ferrous ammonium sulfate, mercuric sulfate, sulfuric acid, ferroin indicator, sodium thiosufate, Winkler's reagent, $MnSO_4$, potassium iodide, starch indicator, citric acid, HCl, EDTA, NaOH, hexane and sodium metabisulphite (SMBS) were purchased from sd Fine Chemicals Ltd., Hyderabad, India. Trimesoyl chloride, piperazine and meta-phenylenediamine were obtained from Sigma Aldrich, Bangalore, India. BOD incubator (RCI-S.NO-313, India), COD analyzer (DRB 200 COD Reactor, Germany) for determination of biochemical and chemical oxygen demand besides Colorimeter (Hach-DR-890) for turbidity analysis were procured from M\s Hach, Bangalore, India. Conductivity meter (DCM-900) and pH meter (DPH-504) were purchased from Global Electronics, Hyderabad, India.

2.2. Synthesis of functionalized NF and TFC polyamide RO membranes

Polyethersulfone (PES) ultraporous substrate of approximately 50 kDa molecular weight cutoff (MWCO) was prepared by phase inversion method using 15% w/v solution of the polymer in dimethyl formamide (DMF) solvent containing 3% v/v propionic acid [18,19]. The homogenous bubble free solution was then cast on a nonwoven polyester fabric support affixed onto a clean glass plate using a doctor's blade followed by immersion in ice cold water bath to obtain ultraporous substrate. To obtain polyamide NF membrane by interfacial polymerization, PES substrate was soaked in 1% aqueous solution of piperazine for 1 min. After draining off excess water, the substrate was immersed in hexane bath containing 0.1% of trimesoyl chloride (TMC) for 30 s. The membrane was then heated in an oven at 110 °C for 10 min to obtain a NF membrane of about 600 MWCO. 0.75% dilute polyvinylalcohol (PVA)

Table 1
Feed characteristics of bulk drug industrial effluent.

Feed characteristics	
TDS (ppm)	5710
Conductivity (mS/cm)	8.45
Turbidity (FAU)	318
Color (Pt–Co)	60
pH	6–6.5
BOD (ppm)	8.409
COD (ppm)	4050